Phase transitions in systems of hard rectangles with non-integer aspect ratio

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Abstract –We investigate, using Monte Carlo simulations, the phase diagram of a system of hard rectangles of size $m \times mk$ on a square lattice when the aspect ratio k is a non-integer. The existence of a disordered isotropic phase, a nematic with only orientational order, a columnar phase with orientational and partial translational order, and a high density phase with no orientational order is shown. The high density phase is a solid-like sublattice phase only if the length and width of the rectangles are not mutually prime, else, it is an isotropic phase. The minimum value of k beyond which the nematic and columnar phases exist are determined for $m = 2$ and 3. The nature of the transitions between different phases is determined, and the critical exponents are numerically obtained for the continuous transitions.

Understanding the nature of the different phases and the transitions between them in a system of hard rods has significance for more complex physical systems such as liquid crystals [1], tobacco mosaic virus [2], fd virus [3–5], silica colloids [6,7], boehmite particles [8,9], DNA origami nanoneedles [10], and adsorption of gas particles on metal surfaces [11–15]. In three dimensional continuum, the system of hard rods undergoes an entropy driven phase transition from a low density isotropic phase to a high density nematic phase that has orientational order [16–18]. Further increase in density may result in a smectic phase with orientational and partial translational order and a solid phase [19]. In two dimensions, the system undergoes a Kosterlitz Thouless type phase transition from an isotropic phase to a power law correlated phase [20]. Hard cuboids on a cubic lattice and hard rectangles on a square lattice are the corresponding lattice analogues. While the complete phase diagram for cuboids is not known, the system of hard rectangles has a rich phase diagram [21].

Consider a system of monodispersed hard rectangles of size $m \times mk$ ($k > 1$) on a square lattice, interacting only through excluded volume interaction. When $m = 1$ (hard rods), the system undergoes two transitions with increasing density for aspect ratio $k \geq 7$: first from a low density disordered phase to an intermediate density nematic phase [22–26], and second, from the nematic phase to a high density disordered phase [27, 28]. The high density disordered phase has been argued to be a reentrant low density disordered phase [29]. When $k = 2$ (dimers), it may be rigorously shown that there are no transitions at non-zero densities of vacancies [30–33], though at full packing, the correlations decay algebraically on bipartite lattices [34], and exponentially on non-bipartite lattices [35]. For $k \gg 1$, the existence of the nematic phase at intermediate densities may be proved rigorously [23]. The only exact solutions that exist are for the model of hard rods on a Bethe-like lattice [29, 36].

For $m \geq 2$ and integer k, four different phases have been observed: isotropic, nematic, columnar, and solid-like sublattice phases [21,37,38]. The nematic phase exists only for $k \geq 7$ for $m = 2, 3$. The columnar phase exists only when $k \geq 4$ for $m = 2$ and for $k \geq 2$ for $m = 3$. For large enough k, with increasing density, the system transits successively from isotropic to nematic to columnar to sublattice phase. The nature of the phase transitions has also been studied. The isotropic-nematic transition belongs to the Ising universality class for all m. When $m \geq 3$, all other transitions are first order in nature. When $m = 2$, all transitions ex-

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cept the isotropic-columnar transition are continuous and belong to the Ising or Ashkin-Teller universality class. The isotropic-columnar transition is continuous for $k = 5$ and first order for $k = 6$, implying the existence of a tricritical point at an intermediate value of k [21].

All the above results are for integer values of the aspect ratio k. Within viral expansion and Bethe approximation, the results do not depend on whether k is an integer or not [37]. However, these results are presumably valid only for large k. For smaller k, when the applicability of the model for adsorption of gas particles on metal surfaces is more relevant, it is not clear what the effect of k being an non-integer is. What are the different phases and the phase diagram when k is rational but not an integer? What are the minimum values of k beyond which the nematic and columnar phases exist? Answering these questions will allow us to obtain the complete phase diagram for the system of hard rectangles. In this paper, we obtain the phase diagram for $m = 2$, when k is a half-integer, using large scale Monte Carlo simulations. The isotropic-columnar transition is shown to be discontinuous while the isotropic-nematic and nematic-columnar transitions are shown to be continuous. The critical exponents are numerically estimated for the continuous transitions. We find that the columnar phase exists only when $k \geq 11/2$ for $m = 2$ and when $k \geq 13/3$ for $m = 3$. The nematic phase exists only when $k \geq 15/2$ for $m = 2$ and when $k \geq 22/3$ for $m = 3$.

The Model and Monte Carlo algorithm. – Consider monodispersed hard rectangles of size $m \times mk$ on a square lattice of size $L \times L$ with periodic boundary conditions. Each rectangle is oriented either horizontally or vertically. A horizontal (vertical) rectangle occupies mk lattice sites along x (y)-direction and m lattice site along y (x)-direction. Each site may have at most one rectangle passing through it. We associate an activity $z = e^{\mu}$ to each rectangle, where μ is the chemical potential. In this paper, we restrict the aspect ratio k to non-integers.

We simulate the system in the constant μ grand canonical ensemble using an efficient algorithm involving cluster moves that has been shown to be very useful in equilibrating hard core systems of extended particles at high densities [21, 27, 28]. Here we briefly review the algorithm. Starting from a valid configuration, a row or a column (say a row) is chosen at random. All horizontal rectangles whose bottom-left corners (heads) are on that row are evaporated, keeping the rest of the configuration unchanged. The row now consists of intervals of empty sites, separated by sites that are either occupied by rectangles or can not be occupied due to the hard core constraint. The empty intervals of the row are reoccupied by a new configuration of horizontal rectangles with the correct equilibrium grand canonical probabilities. The calculation of these probabilities reduces to a solvable one-dimensional problem. If a column is chosen, similar evaporation-deposition moves are performed for vertical rectangles. Equilibration is faster on including a flip move in which a square plaquette consisting of ℓ aligned horizontal or vertical rectangles is rotated by $\pi/2$, where ℓ is the ratio of the least common multiple of m and mk to m. A detailed description of the implementation of the algorithm for the system of hard rectangles is described in Ref. [21]. Unlike for integer k, the flip move is less effective for non-integer k because the rotatable plaquettes are larger in size and hence, have lower probability to occur during the simulations. This makes it difficult to equilibrate the systems at high densities. Other implementations of the algorithm include lattice models of hard rods [27, 28], hard discs [39], and mixtures of dimers and hard squares [40].

Different Phases. – As for integer k, we observe four different phases in the simulations: an isotropic (I) phase, a nematic (N) phase, a columnar (C) phase and a high density (HD) phase. The I phase is disordered. In the N phase, rectangles orient preferably along the horizontal or vertical direction, but they do not have any positional order. Each row or column on an average contains equal number of heads of rectangles. The columnar (C) phase has orientational order and translational order only in the direction perpendicular to the nematic orientation. When $m = 2$, in the columnar phase, if the majority of the rectangles are horizontal (vertical), their heads lie mostly on even rows (columns) or odd rows (columns). Hence, there are 4 (in general $2m$) symmetric C phases. The I, N and C phases are observed when $m \geq 2$, for both integer and non-integer k.

The HD phase has no orientational order. But it may or may not possess translational order depending on the length and width of the rectangles. Let, the greatest common divisor of the length and width be denoted by p. We divide the square lattice into p^2 sublattices by assigning to a site (i, j) a label $(i \mod p) + p \times (j \mod p)$. In the fully packed limit, it is straightforward to verify that the heads of the rectangles occupy one of the p^2 sublattices. We expect this phase to be stable to introduction of vacancies at densities close to the full packing. If $p > 1$, the HD phase is a sublattice phase with complete translational order but no orientational order [21]. On the other hand, when $p = 1$ (length and width are mutually prime), the HD phase is disordered with no orientational or translational order. Since existing evidence for $m = 1$ suggests that the high density disordered phase is qualitatively similar to the low density I phase [28, 29], we expect the same to hold for $m \geq 2$ whenever the HD phase is disordered.

When $m \geq 2$ and integer k, $p = m > 1$ and the HD is known to be a sublattice phase [21], consistent with the above argument. To further confirm that the HD phase is a sublattice phase when $p > 1$, but k is a non-integer, we simulate the system of rectangles of size 4×6, for which $p = 2$. We divide the lattice into $p^2 = 4$ sublattices. The sublattice order parameter is defined as $q_1 = n_0 - n_1 - n_2 + n_3$, where n_i is the fraction of sites oc-

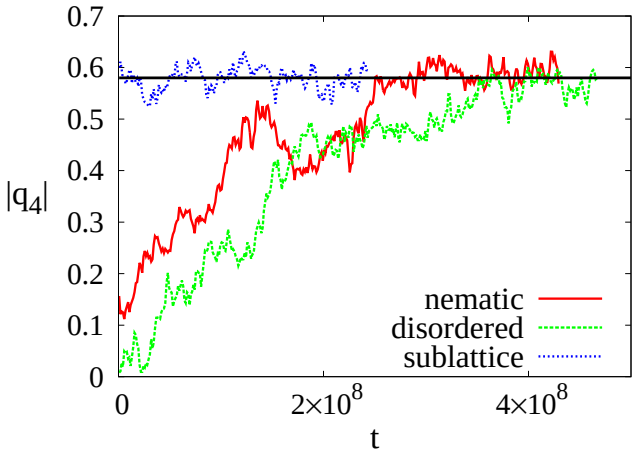

Fig. 1: Time evolution of the sublattice order parameter q_1, starting from three different types of initial configurations: nematic, disordered, and sublattice phases. The straight line is $|q_1| = 0.580$. The data are $\mu = 10.8$ and for L=960. The equilibrium density is ≈ 0.962.

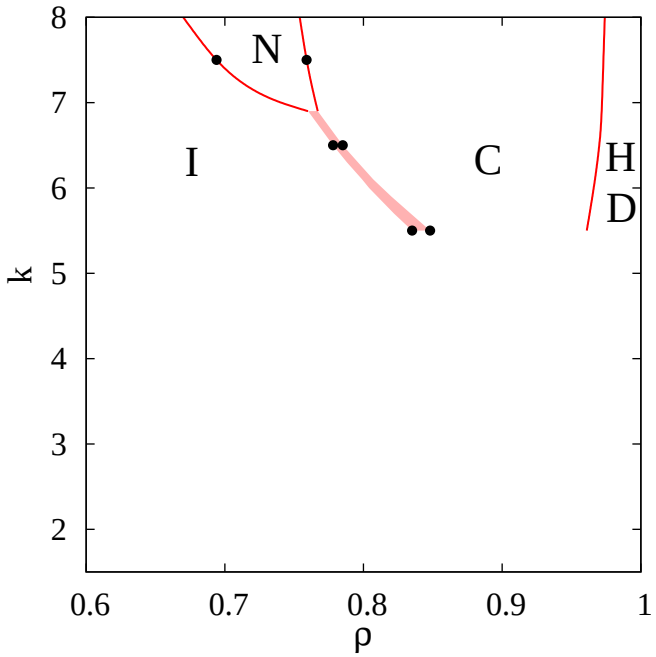

Fig. 2: Phase diagram for rectangles of size $2\times 2k$, where k is restricted to non-integer values. I, N, C and HD denote isotropic, nematic, columnar and high density phases respectively. The data points are from simulation, while the continuous lines and shaded portions are guides to the eye. The shaded portion denotes regions of phase coexistence.

cupied by rectangles whose heads are on the ith sublattice. It is straightforward to check that $\langle|q_1|\rangle \neq 0$ only for the sublattice phase. To show the existence of sublattice phase at high density, a large value of μ is chosen ($\mu = 10.8$), and the temporal evolution of $|q_1|$ is tracked, starting from three different initial configurations: nematic, disordered and sublattice phases (see Fig 1). At large times, the system reaches a stationary state that is independent of the initial configuration, ensuring equilibrium. For this choice of μ, the fraction of occupied sites ρ fluctuates around 0.962. In equilibrium, $\langle|q_1|\rangle \approx 0.580$, clearly showing the existence of a sublattice phase.

Phase Diagram for $m = 2$. – The phase diagram for $m = 2$ and non-integer k is shown in Fig. 2, where the data points are obtained from Monte Carlo simulations and the lines, based on analysis of the phase diagram for large k [37] are guides to the eye. The low density phase is an I phase for all k. Since the length and width of the rectangles are mutually prime, $p = 1$, and the HD phase is a reentrant I phase. No phase transitions are observed when $k \leq 9/2$. The C phase exists only for $k \geq 11/2$, while the N phase exists only for $k \geq 15/2$.

We could not numerically obtain any data point on the C-HD phase boundary as it is not possible to equilibrate the systems within available computer time at high densities for $k \geq 11/2$. However, the critical density for C-HD phase transition was argued to behave asymptotically as $1 - a/(mk^2)$, for $k \gg 1$, where a is a constant. Likewise, for large k, the critical density for the I-N phase transition scales as Ak^{-1}, where A is a constant, independent of m, and that for the N-C phase transition tends to a non-zero constant [21, 37]. The solid lines in Fig. 2 follow these asymptotic behavior for large k.

The I-C transition is found to be first order for both $k = 11/2$ and $13/2$. The shaded region in Fig. 2 denotes the region of phase coexistence at a first order phase transition. We find that the I-N and N-C transitions are both continuous. These transitions are analyzed in detail below.

Critical behavior for $m = 2$. – We now study the nature of the different phase transitions for the system of $2 \times 2k$ rectangles, where k is half-integer. To study the I-N transition, we define the order parameter

$$q_2 = n_h - n_v, \tag{1}$$

where n_h and n_v are the fraction of sites occupied by the horizontal and vertical rectangles respectively. In the I phase $\langle|q_2|\rangle = 0$, while in the N phase, $\langle|q_2|\rangle \neq 0$.

The I-C and N-C phase transitions are best studied with the order parameter

$$q_3 = |n_{re} - n_{ro}| - |n_{ce} - n_{co}|, \tag{2}$$

where n_{re} (n_{ro}) is the fraction of sites occupied by rectangles whose heads are in the even (odd) rows, and n_{ce} (n_{co}) is the fraction of sites occupied by rectangles whose heads are in the even (odd) columns. In the I and N phases, $n_{re} \approx n_{ro}$, and $n_{ce} \approx n_{co}$, implying that $\langle|q_3|\rangle = 0$. In the C phase, either $n_{re} \neq n_{ro}$ and $n_{ce} \approx n_{co}$, or $n_{ce} \neq n_{co}$ and $n_{re} \approx n_{ro}$ implying that $\langle|q_3|\rangle \neq 0$.

The other relevant thermodynamic quantities are the second moment χ_i and the Binder cumulant U_i, defined

as

$$U_i = 1 - \frac{\langle q_i^4 \rangle}{3\langle q_i^2 \rangle^2}. \quad (3a)$$

$$\chi_i = \langle q_i^2 \rangle L^2, \quad (3b)$$

where $i = 2, 3$. Near the critical point, the singular behavior is captured by finite-size scaling:

$$\langle |q_i| \rangle \simeq L^{-\beta/\nu} f_q(\epsilon L^{1/\nu}), \quad (4a)$$

$$U_i \simeq f_u(\epsilon L^{1/\nu}), \quad (4b)$$

$$\chi_i \simeq L^{\gamma/\nu} f_\chi(\epsilon L^{1/\nu}), \quad (4c)$$

where $\epsilon = (\mu - \mu_c)/\mu_c$, where μ_c is the critical chemical, β, γ, ν are the critical exponents, and f_q, f_u, and f_χ are scaling functions.

Isotropic–Nematic (I-N) transition. We study the I-N transition for 2×15 ($k = 15/2$) rectangles using the order parameter q_2. Since the N phase may have orientational order only in the horizontal or vertical direction, we expect the I-N transition to be in the two-dimensional Ising universality class, as has been confirmed for integer k, when $m = 1$ [24] and $m = 2, 3$ [21], and for systems of polydispersed rods [41, 42]. The data for U_2 for different system sizes intersect at $\mu = \mu^c_{I-N} \approx 0.945$ [see Fig. 3(a)]. The corresponding critical density is $\rho^c_{I-N} \approx 0.694$, which is less than $\rho^c_{I-N} \approx 0.745$ for $k = 7$ [28], consistent with $\rho^c_{I-N} \approx Ak^{-1}$ [37]. The data for U_2 [see Fig. 3(b)], $\langle |q_2| \rangle$ [see Fig. 3(c)], and χ_2 [see Fig. 3(d)] for different system sizes collapse onto a single curve when scaled as in Eq. (4) with Ising exponents $\beta/\nu = 1/8$, $\gamma/\nu = 7/4$, and $\nu = 1$. For larger values of k, integer or otherwise, we expect the I-N transition to be in the Ising universality class.

Nematic–Columnar (N-C) transition. We study the N-C transition for rectangles of size 2×15 using the order parameter q_3. When the system makes a transition from the N phase with horizontal (vertical) orientation to the C phase, the symmetry between even and odd rows (columns) is broken. From symmetry considerations, we expect the N-C transition to be in the Ising universality class. The data for U_3 for different system sizes intersect at $\mu = \mu^c_{N-C} \approx 1.696$ [see Fig. 4(a)], corresponding to $\rho^c_{N-C} \approx 0.759$. The data for U_3 [see Fig. 4(b)], $\langle |q_3| \rangle$ [see Fig. 4(c)], and χ_3 [see Fig. 4(d)] for different system sizes collapse onto a single curve when scaled as in Eq. (4) with Ising exponents $\beta/\nu = 1/8$, $\gamma/\nu = 7/4$, and $\nu = 1$. The N-C transition in the system of 2×14 rectangles has also been shown to be in the Ising universality class [21], and we expect the same for $k > 15/2$.

Isotropic–Columnar (I-C) transition. The I-C transition occurs only for rectangles of size 2×11 ($k = 11/2$) and 2×13 ($k = 13/2$). The transition is best studied using the order parameter q_3. We find that the transition is first order for both values of k. This may be established by numerically calculating the probability density functions (pdf) $P(\rho)$ of the density ρ and $P(q_3)$ of the order parameter q_3 for values of μ that are close to the transition

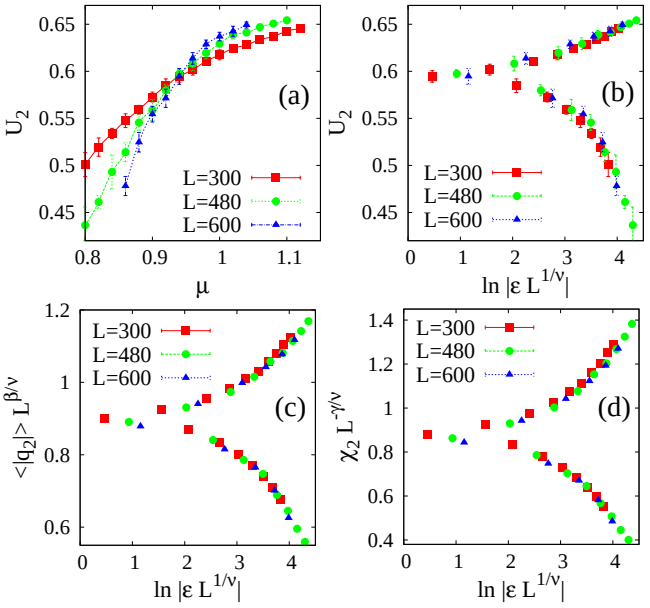

Fig. 3: The critical behavior near the I-N transition for rectangles of size 2×15 ($k = 15/2$). (a) The data for Binder cumulant for different system sizes intersect at $\mu^c_{I-N} \approx 0.945$ ($\rho^c_{I-N} \approx 0.694$). The data for different L near the I-N transition for (b) Binder cumulant, (c) order parameter, and (d) second moment of the order parameter collapse onto a single curve when scaled as in Eq. (4) with the Ising exponents $\beta/\nu = 1/8$, $\gamma/\nu = 7/4$, and $\nu = 1$.

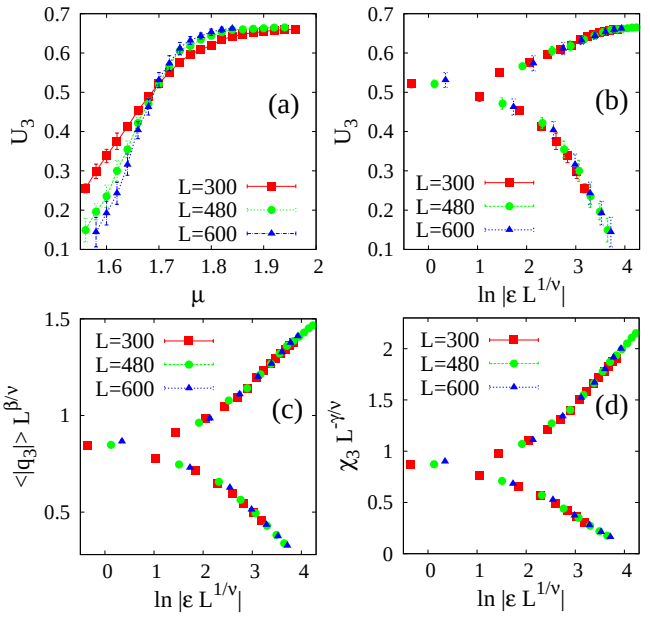

Fig. 4: The critical behavior near the N-C transition for rectangles of size 2×15 ($k = 15/2$). (a) The data for Binder cumulant for different system sizes intersect at $\mu^c_{N-C} \approx 1.696$ ($\rho^c_{N-C} \approx 0.75$). The data for different L near the N-C transition for (b) Binder cumulant, (c) order parameter, and (d) second moment of the order parameter collapse onto a single curve when scaled as in Eq. (4) with the Ising exponents $\beta/\nu = 1/8$, $\gamma/\nu = 7/4$, and $\nu = 1$.

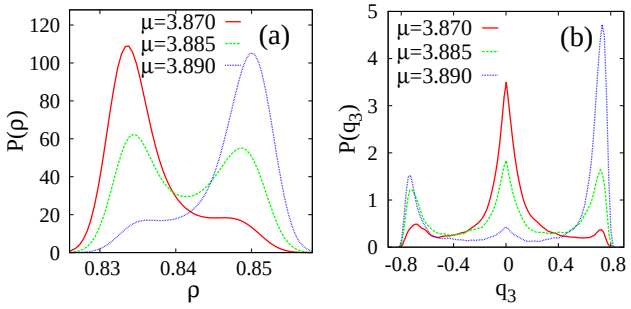

Fig. 5: Probability density function of (a) density ρ and (b) order parameter q_3 for three values of μ near the I-C transition. The data are for rectangles of size 2×11 and $L = 440$.

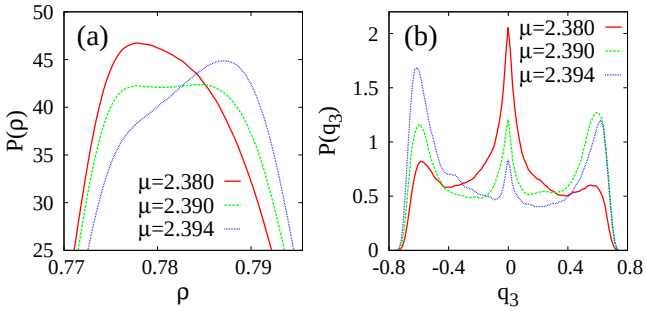

Fig. 6: Probability density function of (a) density ρ and (b) order parameter q_3 for three values of μ near the I-C transition. The data are for rectangles of size 2×13 and $L = 416$.

point $\mu^c_{I-C} \approx 3.885$ for $k = 11/2$ and $\mu \approx \mu^c_{I-C} = 2.390$ for $k = 13/2$. The pdf for $k = 11/2$ and $13/2$ are shown in Fig. 5 and Fig. 6 respectively. In both the figures, the pdfs for q_3 have three clear peaks at the transition point: the two peaks at $q_3 \neq 0$ correspond to the symmetric C phases and the one at $q_3 = 0$ to the I phase. The pdf for ρ have two peaks of nearly equal height at the transition point, though these peaks are not clearly separated for $k = 13/2$. The difference in the peak positions is equal to the jump in the density across the transition point and is shown by the shaded region in Fig. 2. We find that these peaks become sharper with increasing system size. These are clear signatures of a first order transition.

Phase Diagram for $m \geq 3$. – We expect that the phase diagram for $m \geq 3$ and non-integer k to be qualitatively similar to that for $m = 2$ with three entropy driven transitions for large k. The HD phase is a disordered or sublattice phase depending on whether the length and width of the rectangles are mutually prime or not. For $m = 3$, we determine the the minimum value of k beyond which the C and N phases exist. There are no transitions for $k \leq 11/3$. We find that the C phase exists only for $k \geq 13/3$, while the N phase exists only for $k \geq 22/3$. When $m = 3$, the C phase has a 6-fold symmetry, and thus with analogy with Potts model, we expect a first order transition. For rectangles with $k = 13/3$, we confirm that the I-C phase transition is first order in nature. The pdfs for ρ and q_3 behave similarly to that for the case of $m = 2$. All the other transitions are also expected to be first order except the I-N transition, as seen for integer k [21].

Summary and Discussions. – In this paper, we obtained numerically the phase diagram of the system of hard rectangles of size $m \times mk$ with non-integer aspect ratio k. As for integer k, the system may exist in four different phases: isotropic, nematic, columnar or high density phase. For integer k, the high density phase is a solid-like sublattice phase. However, when k is a non-integer, the high density phase is a disordered phase when the length and width of the rectangles are mutually prime. The phase diagram for large m is expected to be qualitatively similar to that for $m = 2$. The isotropic-nematic transition will be in the Ising universality class for all m. However, all other transitions are expected to be first order.

The N phase is found to exist only when $k \geq 15/2$ for $m = 2$ and when $k \geq 22/3$ for $m = 3$. For integer k, the N phase exists only when $k \geq 7$ for $m = 1, 2, 3$ [21,22]. These different lower bounds may be combined to give tighter bounds for k^{I-N}_{min}, the smallest value of k beyond which the N phase exists. We conclude that $20/3 < k^{I-N}_{min} \leq 7$.

The bounds for k^{I-C}_{min}, the minimum value of k beyond which the C phase exists, are not so clear. We find that the C phase exists when $k \geq 11/2$ for $m = 2$ and when $k \geq 13/3$ for $m = 3$. On the other hand, for integer k, the C phase exists for $k \geq 4$ for $m = 2$ and when $k \geq 2$ for $m = 3$. Thus, unlike for the N phase, k^{I-C}_{min} depends both on m and whether k is a integer or not, and it is not possible to combine the bounds only in terms of m.

The phase diagram when the aspect ratio of the rectangles is irrational remains an open question. In this case, it has been conjectured that there could be more transitions at densities close to full packing, when the disordered phase will become unstable to a nematic or columnar phases [22]. This question, as well as finding tighter bounds on k for existence of different phases are best answered by obtaining the phase diagram of hard rectangles with restricted orientation in two dimensional continuum, when the aspect ratio may be continuously tuned.

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The simulations were carried out on the supercomputing machine Annapurna at The Institute of Mathematical Sciences.

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